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Indirect Additive Manufacturing of resin components using polyvinyl alcohol sacrificial moulds

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Abstract

Using additive manufacturing (AM) as aid to manufacture parts with traditional manufacturing methods is one of the many applications of AM, yet one of the less investigated. In this article, for the first time, a way to produce soluble AM customizable sacrificial moulds for resin casting is explained. The moulds are produced through fused filament fabrication using polyvinyl alcohol (PVA) as raw material. After curing, the moulds are dissolved in water leaving the solid resin parts away. The results of indirect additively manufactured resin components in PVA moulds are examined, supported by surface and dimension analysis on prism-like sample parts for different sets of process parameters. Possible applications and limitations of the technique are exposed, as well as recommendations for future works.

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1. Introduction

The evolution of the industry towards smart and tailored products increases the necessity of mass customization, this is a big drive to develop new non-traditional manufacturing methods. Hence, AM, commonly referred as 3D printing, is considered a key technology to aid the so called fourth industrial revolution [1]. This is because of its ability to create complex objects in terms of shape and material. Many applications have exploited the benefits of AM [2], but many other applications are to be discovered, as it is an emerging manufacturing technology. Normally when the public thinks about 3D printing, it is to directly fabricate parts, i.e. to have the desired object straight out of the printer. Thus, there is a lack on the research of indirect manufacturing methods, where AM is used as aid to produce objects with more traditional techniques. The need of using Indirect Additive Manufacturing (I-AM) raises, when classic AM cannot handle some specific material, when isotropic mechanical properties are needed, or

for simply expanding the design freedom that direct AM restricts, because of its inherent manufacturing constraints [3].

In this article the possibilities of creating customizable soluble moulds for resin casting are explored, supported by experiments and surface analysis. Epoxy resins are widely investigated and are the backbone of the composite materials[4], an area with many years of development compared to the short period of AM materials. The remarkable properties that can be reached using epoxy resins, plus the opportunity of rapid creating precise intricate shapes, enable the designers to deploy new structures in shorter periods of time. In this way it is possible to achieve lightweight parts with good mechanical behaviour.

About direct AM, several studies on design guidelines and recommendations have been published [5]–[7]. For I-AM those studies are sparser, since depending on the specific application the design rules can change abruptly. The intention of this article is to facilitate a way to the designers to create resin casts with AM sacrificial water-soluble moulds together with some

recommendations based on dimensional and surface analysis. The latter set the basis for the future development of formal design rules in the development of soluble moulds for resin casting. The outcome presented in this article is actually being used by a design team at the Institute for Technical Product Development of the University of the Bundeswehr Munich.

Nomenclature

AM	Additive Manufacturing
I-AM	Indirect Additive Manufacturing
PVA	Polyvinyl Alcohol.
FFF	Fused Filament Fabrication
DfAM	Design for Additive Manufacturing

2. State of the Art

2.1. Additive Manufacturing

The international standard for Additive Manufacturing (AM) provided by the International Organization for Standardization (ISO), defines the term AM as the process of joining materials to make parts from 3D model data, usually layer upon layer, as opposed to subtractive manufacturing and formative manufacturing methods [8]. Additionally, the term 3D printing is defined by the same standards as the fabrication of objects through the deposition of material using a print head, nozzle, or another printer technology. Until recent time 3D printing was used to refer to consumer focus, low-end in price and/or overall capability AM systems, while AM was used for industrial focus high-end AM systems. Because of all the recent popularity and media attention that 3D printing has gained, the term began to be used as synonym of AM. Nowadays 3D printing is the *de facto* standard term [9], and it is the most popular of the two. Even industry leaders use the term 3D printing to promote their high-end AM systems [10].

Originally AM was mostly used for *Rapid Prototyping* [11], i.e. the rapid creation of physical prototypes in an early stage of the product development. During the last years, the technology has reached maturity and is starting to establish itself as an industrial manufacturing technology [12]. Currently, AM is also being used for both *Rapid Direct Manufacturing* and *Rapid Tooling* solutions [13]. This technology is applied in several areas like aerospace, medical, spare parts and tooling [14].

AM is applicable to many materials such as polymers, metals or ceramics and therefore different AM technologies are currently available. Some of the main advantages (in contrast to subtractive manufacturing) are their ability to form complex geometries, the opportunity of on-demand manufacturing and the possibility of creating customized parts in a low volume production. Because of its capabilities and the ease of integration into smart manufacturing digital systems, AM became one of the enablers of Industry 4.0 [1].

2.2. Indirect Additive Manufacturing

AM is normally used to produce functional parts or prototypes in a direct way, i.e. the additively produced part is the object to be used. Nevertheless, AM can be used indirectly in a way of manufacturing master patterns, tools or any means to be used in a non-AM processes to produce final parts. This is called Indirect Additive Manufacturing (I-AM) [15].

I-AM is currently used for the production of lost patterns for casting or vacuum casting, investment casting, sand casting moulds [16], tools for die casting and tools for injection moulding [17], with special interest in the ceramic industry [18]. A novel application is the one presented by Van Hoorick et al. [19], where AM is used to produce a sacrificial polyester scaffold, in order to support hydrogel material and prevent the collapse of a very complex hydrogel structure before UV-curing. Another interesting I-AM example is presented in the publication of Mun et al. [20], where an aluminium alloy is casted into an additively manufactured wax mould with the shape of a cellular structure.

2.3. Fused Filament Fabrication

Fused Filament Fabrication (FFF), commercially referred as Fused Deposition Modelling (FDM[®]), is an AM method that uses a thermoplastic filament as feedstock material. FFF uses an extruder that heats up and forces the thermoplastic filament out, to deposit the semi-molten polymer onto a platform in a layer-by-layer process. The filament is pushed through the extruder by two tractor rollers. At the end of each finished layer, the base platform is lowered by a specific amount, giving the layer thickness, and the next layer is deposited [21]. FFF belongs to the classification *Material Extrusion* of the process categories section of the ISO/ASTM 52900 standards [22].

Sometimes when the extruded material cannot structurally hold by itself, due to the high overhang angle or challenging extrusion conditions, support material is needed. After the printing job, the support structure is taken away by several methods depending on the nature of the support material. Usually it is dissolved in a solution that does not affect the structural material. The material supply is provided in form of a wire filament roll.

In the FFF process the quality or resolution of a part depends on the layer thickness and the nozzle diameter, common values are between 0.1 mm to 0.5 mm. The overall printing time is in relation with those two parameters. Despite the poorer quality for fast production and lower mechanical properties, especially in the out of plane direction, FFF is one of the most common used additive manufacturing technologies for polymers. Its main application is the fast production of prototypes, also called Rapid Prototyping (RP) [23]. Different thermoplastics are used in the FFF process. Among the best known are ABS, PLA, Nylon and Polycarbonate. A schematic of FFF is presented in Fig. 1.

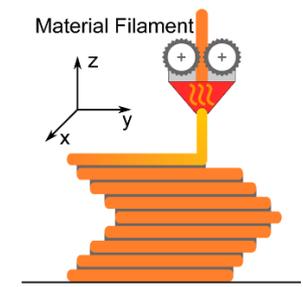


Fig. 1. FFF process schematic [24]

2.4. Polyvinyl Alcohol

PVA, sometimes referred as PVOH is a thermoplastic synthetic polymer. It is biodegradable and highly hygroscopic. When it is exposed to water long enough, PVA dissolves. PVA is produced by hydrolysis of polyvinyl acetate by removal of the acetate groups. Depending on the hydrolysis degree during production, the melting point (T_m) of PVA is between 180 °C (partially hydrolysed) and 220 °C (fully hydrolysed). The lower the degree of hydrolysis and polymerization of PVA, the higher is its solubility in water and the easier is its crystallization [25]. The glass transition temperature (T_g) is 85 °C and the degradation temperature ranges between 350 – 450 °C.

PVA is currently used in the food packaging industry, water treatment, textile, agriculture, cleaning and detergent products, as additives in construction and in medical devices. PVA is also used as a raw material for AM, mainly as support structure for the material extrusion methods, due to its relatively good mechanical properties coupled with the convenience that the material dissolves under water. PVA has also proven to be successful in AM of tailored pharmaceuticals due to its biodegradability [26], using the material as carrier of drugs through inkjet methods.

2.5. Epoxy Resin

Epoxy resins are a family of monomeric or oligomeric material that can be further reacted to form thermoset polymers [27]. Their reactivity enables them to bond well to fibres and their toughness. Normally they are combined in liquid state with glass, carbon, or aramid fibres, to cure and produce solid composite materials with the best properties of most thermosets [28]. Many uses of this material have been exploited since it has been under intense research since the 1950s [29]. Among the most known applicants are for example the aerospace, medical and automotive industry.

Epoxy resins utilized as well for direct AM. Commonly acting as binding agents or in the case of being photocurable, they are used to build objects through photopolymerization [30]. Furthermore, it is possible to use them as binding agents for composites, for example with embedded magnetite particles for electromagnetic shielding [31] or continuous carbon fibre composites [32].

3. Implementation

In this study the manufacturing of resin parts using FFF PVA moulds is analysed. The mould is created from the negative design of the part. If DfAM guidelines and recommendations were existent for this application of I-AM, this design would be enriched with them. Ideally, the production of the resin part would involve the design of the part & mould, the AM of the mould, the casting of the resin, followed by the curing and dissolving of the mould, like it is shown in the process scheme of Fig. 2.

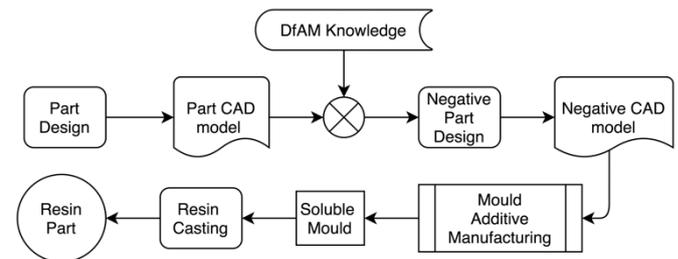


Fig. 2. I-AM process schematic

When designing for I-AM the DfAM guidelines and recommendations apply on the manufacturing aid or tooling and not directly on the designed part. In this case DfAM would apply on the mould design. Since the design of soluble PVA moulds for resin casting is not actually covered by DfAM rules, in this section a test specimen is designed and manufactured with the aim of generating recommendations for future design applications. In those specimens, deviations and surface roughness are evaluated.

In order to test the manufacturing deviations of the resin parts produced by FFF I-AM, a rectangular prism is analysed as a simple test-part with different manufacturing parameters. To use I-AM for this part, it is necessary to produce an artefact that acts as a mould. The mould is designed in the CAD software considering no deviations with the part. The mould is referred as *artefact* and the resulting resin part as *part*.

After the artefact is produced by FFF in PVA the resin is poured into it. Then, the resin is cured and the soluble artefact is washed away with water, leaving the solid resin part out. As a last step a dimensional analysis is carried out on the part and is compared to the designed dimensions of the artefact. The surface roughness is also measured and evaluated on the artefact and on the part, to see how the imprint is transferred. This process is displayed in Fig. 3.

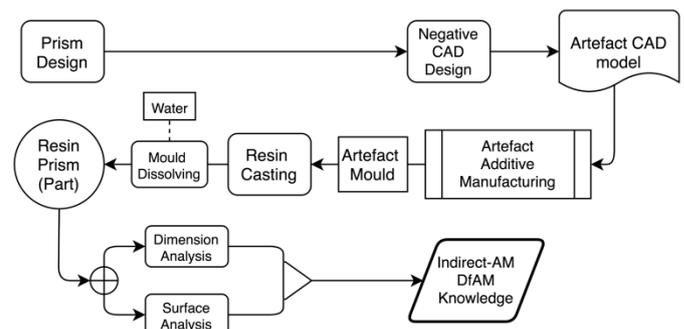


Fig. 3. Implementation process schematic

3.1. Artefact fabrication

The mould artefact is produced through FFF with the material PVA. Both, machine and material are of the brand Ultimaker®. To evaluate the effect of the printing resolution on the resin parts, the surface of the soluble moulds is analysed. Two manufacturing parameters are varied in the production of 6 different types of artefacts. The parameters under variation are material extrusion temperature and layer thickness, according to Table 1. These parameters are chosen due to their known effect on the surface quality [33] for the FFF technology. The range of variations of the layer thickness is taken from the capabilities of the used FFF machine, and the extrusion temperature from the recommendations of material manufacturer.

A batch of 24 artefacts is produced, 4 of each type. For all the artefacts the printing speed is 35mm/s, the infill density is 50% and the building plate temperature is 60 °C (recommended by the material manufacturer). The parameters under variation are material extrusion temperature and layer thickness, according to Table 1.

Table 1. Artefact manufacturing parameters

Specimen	Extrusion Temperature (°C)	Layer Thickness (mm)
Type I	215	0.15
Type II	215	0.10
Type III	215	0.06
Type IV	225	0.15
Type V	225	0.10
Type VI	225	0.06

The geometric design and dimensions of the artefact can be observed in Fig. 4 together with a picture of one produced artefact.

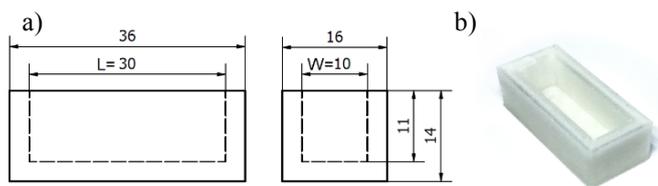


Fig. 4. Artefact dimensions (a) and produced artefact (b).

3.2. Part fabrication

By knowing the density of the resin and hardener blend, 3cm³ of the liquid mix is poured into the artefact using a high precision scale. The sample is cured 24h at 25°C in a room with 5% of relative humidity to avoid PVA detriment. The used resin is Epoxy L and the hardener is L from the brand R&G Faserverbundwerkstoffe® Composite Technology. The cured resin in the artefact container can be seen in Fig. 5 (a).

As last step the artefact is vanished away with water. To accelerate the process the PVA removal is carried out under water at 50°C for a period of 5 hours in an ultrasonic cleaner. The resulting part can be observed in Fig. 5 (b).

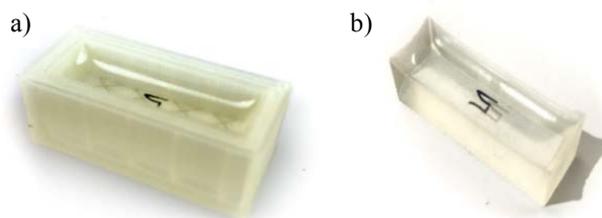


Fig. 5. Resin cured in container (a) & resulting resin part (b)

4. Results & Analysis

4.1. Dimensional deviations

To evaluate possible deviations, the geometrical features of the artefact and part are checked and compared with the designed geometries. The produced resin rectangular prism has two main geometrical parameters: length (L) and width (W). It makes no sense to measure any other dimension, since the surface tension, created by the viscosity of the liquid resin, makes it adhere to the wall of the mould, making the upper surface quasi-convex instead of plane.

The parameters L and W of Fig. 4, are reported for both, the part and artefact in Table 2. As it can be seen, the lengths L and W are shorter than the designed value on the PVA artefact. This happens because the extruded PVA expanded, so the walls of the artefact that supposed to be of 3mm were thicker in all the samples, making the pool smaller in both dimensions.

The resin parts show additional shortening in the L dimension (except for type IV sample), and expansion in the W dimension. It is known that resin shrinks during the curing process [34], so the shortening in L is expected. Regarding the expansion in W, might be because a possible Poisson effect coupled to an uneven curing rate, forcing the smaller length to expand due to the bigger shrinkage of the bigger length. Overall, said expansion is not sufficient to compensate the previous distortion induced by the manufacturing of the PVA mould in order to obtain the desired W=10mm length. In the case of the longer dimension, the desired value of L=30mm is far away from the real parts, being the shortening of 0.25mm in the worst case, by cause of the shortage of the mould coupled with the shrinkage of the resin.

Table 2. Artefact manufacturing parameters in mm

Specimen	Artefact L	Part L	Artefact W	Part W
Designed	30	30±ΔL	10	30±ΔW
Type I	29.85	29.85	9.83	9.92
Type II	29.76	29.74	9.82	9.92
Type III	29.80	29.75	9.88	9.97
Type IV	29.64	29.71	9.75	9.82
Type V	29.73	29.70	9.76	9.82
Type VI	29.77	29.75	9.87	9.97

4.2. Surface roughness

An important aspect considered in this work is the imprint left by the mould after being washed away. To evaluate this, a surface analysis with a confocal laser microscope Keyence VK-X200 is performed.

In first instance the analysis is performed in the PVA moulds. A picture of an artefact type I after being produced is shown in Fig. 6. There it is possible to clearly see the extruded filaments. A surface analysis as 3D map of the same sample appears in Fig. 7.

To assess the impact of the process parameters mentioned in Table 1, an analysis in multiple points (5 equidistant squared sections of $285\mu\text{m} \times 285\mu\text{m}$) of each sample is carried out. The following listed values are considered of relevance in this study for the evaluation of the surface roughness.

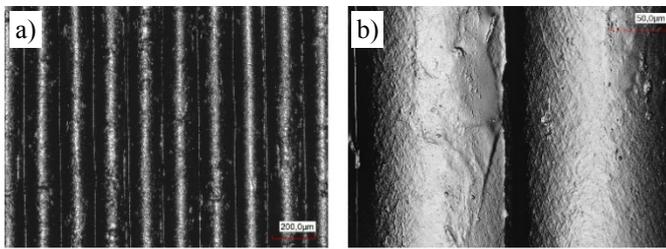


Fig. 6. Artefact type I at 5x (left) & 50x (right) magnification

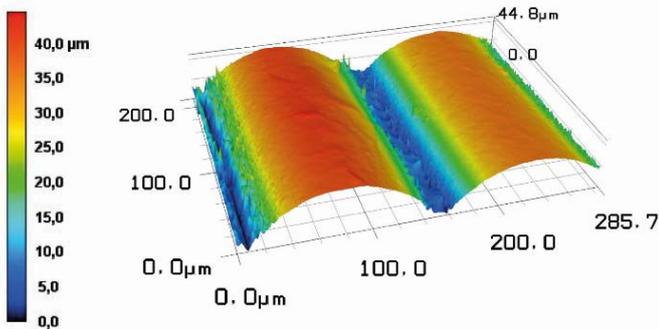


Fig. 7. Roughness 3D map of artefact type I at 50x magnification

- The layer height (“t” in Fig. 8) which is the distance between two consecutive pikes. This value should be coincident with the layer thickness of the manufacturing process.
- A pseudo- R_t , (in this study called simply R_t as in Fig. 8) which is the absolute value of the difference between the highest peak and the lowest valley, in two consecutive similar filaments. Highlighted in yellow in Fig. 8.
- The R_a , arithmetic mean roughness, taking into account the waviness induced by the pile of filaments.

The reported values can be seen in Table 3. Each value is reported as an average of 20 measurements, since a total of 5 measurements are performed in each of the 4 produced samples per type group. In Fig. 9 a picture showing a comparison between a PVA sample type III and VI measurements can be observed. The roughness profile information is taken from the cross-section of this 3D measurement (red line in Fig.9). The cross-section is expanded in Fig.8 for the same samples.

From Fig. 9 it is possible to observe that at the same printing speed and layer thickness, higher extrusion temperatures give a smoother finish by eye, a fact that can be contrasted by the profile analysis of Table 3. The parameter R_a seems to decrease with the increasing extrusion temperature which is a sign of better finishing but the R_t does the opposite. This can be confirmed as it is observed in the magnification of Fig. 8, even though the filaments look glassier in the higher temperature case. They are more irregular in terms of thickness. This gives some really high pikes resulting in a higher value of R_t . Furthermore it can be observed that the valleys are sharper with the increase of the temperature. This is another factor that enlarges the R_t value.

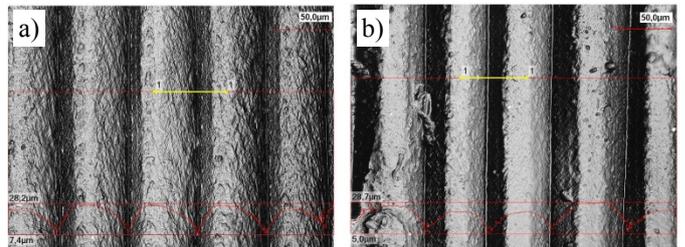


Fig. 9. PVA artefacts type III (a) and VI (b) at 50x magnification

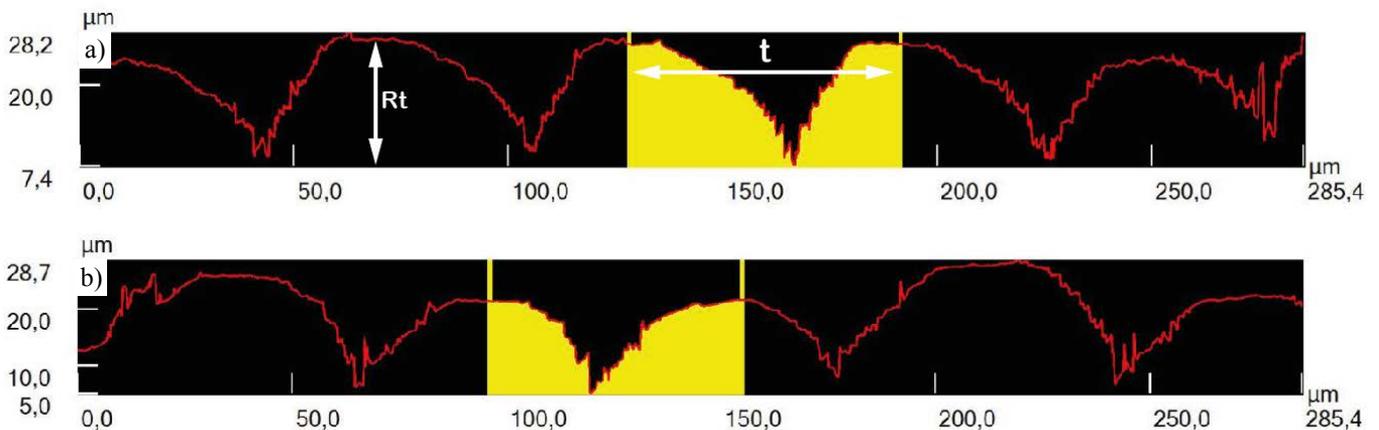


Fig. 8. Roughness profile at 50x, PVA artefact type III (a) and type VI (b)

Table 3. PVA artefact roughness values

Specimen	Layer Height t (μm)	R _t (μm)	R _a (μm)
Type I	151.4	40.7	10.1
Type II	102.9	24.9	7.5
Type III	63.1	19.5	4.4
Type IV	154.6	40.0	10.8
Type V	96.1	33.9	7.6
Type VI	58.8	21.2	3.6

As last instance, the same surface analysis is performed on the resin parts after the mould is vanished away under water. In order to confirm that all the PVA was extracted away, the samples were weighted after washing to check if the cured part matches the weight of the poured resin in section 3.2. Every single weight difference was zero, but some small variations could be lost in the readability of the electronic scale (d=0.001g). These results express that no PVA residue was left in-between the layers, this can be explained by the prolonged ultrasonic cleaning at moderated temperature.

In Fig. 10 a 3D map of the surface of a resin sample type I is displayed. It is possible to see that it is the negative of the one appearing in Fig 5.

The profile analysis is done in the cross-section, analogue to the previous parts. A picture of the laser microscopy is shown in Fig. 12 for samples type III & VI. The reported values follow the same rules as with the PVA artefacts, which can be found in Table 4.

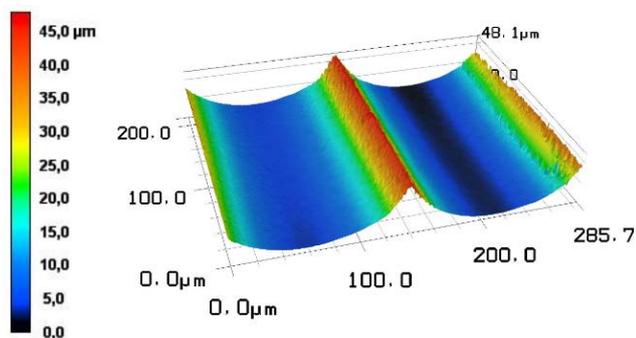


Fig. 10. Roughness 3D map of artefact type I at 50x magnification

Table 4. Resin part roughness values

Specimen	Layer Height t (μm)	R _t (μm)	R _a (μm)
Type I	152.1	45.1	9.4
Type II	102.3	28.7	6.5
Type III	58.2	19.7	4.4
Type IV	147.9	51.9	11.7
Type V	102.9	28.1	6.1
Type VI	59.7	17.5	4.2

As can be seen in Fig. 11, the pikes of this profilometry are decently sharp, which means that the resin filled the bottom of each valley of the PVA mould well. This is confirmed on the cross-section view, which exposes really sharp pikes across the whole measurement. The parameter R_t is following the same pattern as in the PVA mould but the parameter R_a shows a decrement respect to the mould.

The resin parts appear to be less irregular than the PVA parts for the small size defects, as can be observed by comparing Fig. 8 and Fig. 11. A fact that can justify the smaller R_a. Furthermore, can be observed in the comparison of Fig. 12 that the overall bumpy perception of the part type III is less than that in the mould of the same type appearing in Fig. 9.

An interesting fact is that the resin part feels smoother when it is manipulated by hand than the PVA mould for the samples with smaller layer thickness (especially when it is touched in the orthogonal direction to the filaments). This is, because the sharp pikes with the abrupt change of depth do not allow the strong epidermis of the finger to reach the valley of the surface, transmitting a more continuous feeling.

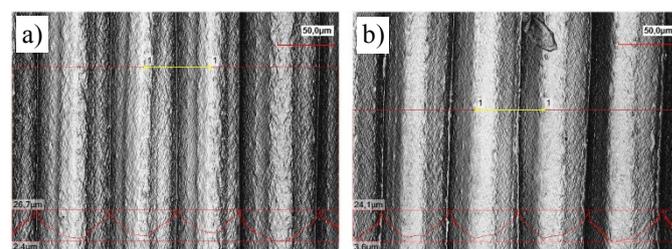


Fig. 12. Resin parts type III (a) and VI (b) at 50x magnification

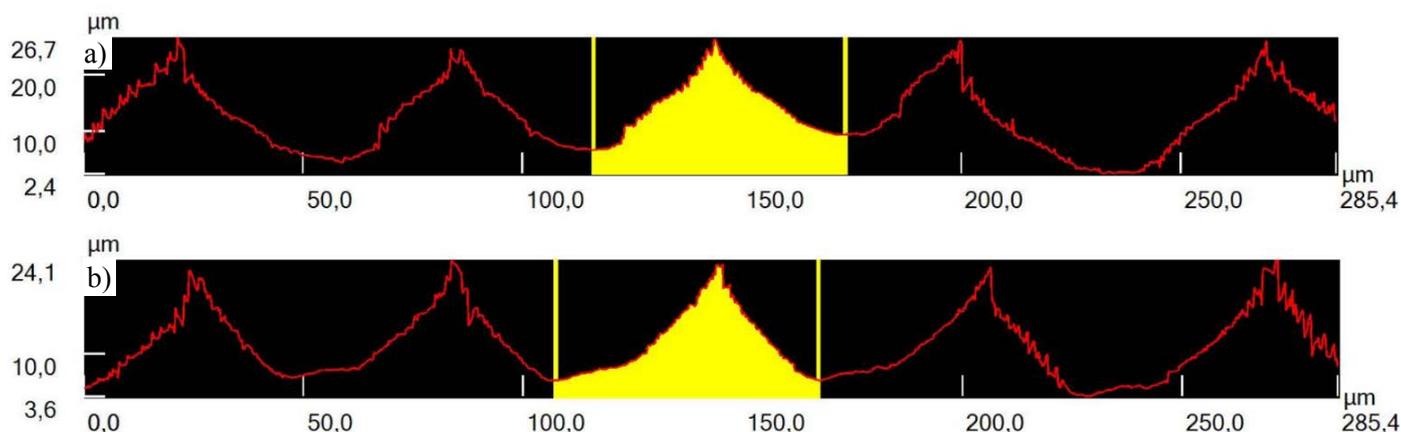


Fig. 11. Roughness profile at 50x, resin part type III (a) and type VI (b)

4.3. Technology applications

This method can be applied to fabricate any sort of resin casted geometry, since the water-soluble PVA mould is not affected by the resin and vice versa. Among the applications that can be exploited from I-AM of resin parts, is the fabrication of high-performance composites with complex shapes. Currently the Institute of Lightweight Structures at the University of the Bundeswehr Munich, is working on the development of this technique to fabricate carbon fibre lattice structures. As opposed to the work of Blattmann et al. [35], who used direct AM to produce octahedral lattice structures with the laser sintering process. This formed a collaboration project with the authors and is subject of future publications.

Another possible application is for some specific parts that require to resist temperature. Using this method is a fast solution instead of using normal FFF with ABS material, since extruded plastics start strength degrading at lower temperatures than epoxy resins [36].

The authors are currently implementing this in the production of spare parts for military applications. This is as well a topic for future publications. Considering the novelty of this I-AM method, many other new applications are on the way to be discovered and investigated.

5. Conclusion

The use of PVA as building material in FFF methods is quite similar to the use of standard materials such as ABS or PLA, but with a slightly higher material expansion after it gets out of the nozzle. This was reflected in the dimensional analysis of section 4.1 and some extra tests of shrinkage of holes in PVA parts. These show that all the holes tend to be smaller than in the CAD file because of the filament expansion. Nevertheless, it is possible to create soluble parts with PVA compensating these deviations in the design. Regarding the manufacturing parameters, the better performing extrusion temperature was 225 °C giving the best surface quality of the set.

The creation of PVA moulds was successfully implemented, the reactivity with epoxy resin is null and the resin copies all the imperfections of the moulds very well. The shrinkage of the resin while curing affects mainly the macro-dimensions but the surface imperfections of the mould are very well traced and transferred to the resin part. The PVA mould vanishes away completely in water, leaving a clean resin part. This was confirmed by the microscope analysis on the resin parts, which showed no residues on the part surface.

The sharp pikes on the resin parts showed no major differences with the valleys of their PVA counterpart. Whenever the specimens are manipulated, the resin part feels smoother or softer to the touch, this it is just a perception effect. This does not mean that the part is smoother than the mould, the microscope analysis and profilometry confirmed that they are actually similar in terms of surface roughness quality.

The presented results and recommendations are the starter to the future development of formal design rules for the creation of PVA moulds for resin casting. This I-AM method opens a wide range of possibilities. The rapid creation of moulds for resin parts is very interesting for such applications,

where normal FFF is limited by the material or the complexity of the object. More applications and use cases of the presented method are to be discovered and investigated.

As future work, the authors aim to develop new uses of this method in the industry. Moulds for the creation of complex composite parts is an interesting application, and would positively affect the rapid design and manufacturing of such components. Furthermore, applications of this method for the aerospace industry are under development at the Institute of Lightweight Structures and the Institute for Technical Product Development at the University of the Bundeswehr Munich. Lastly, the authors encourage the research community to try this method and present the outcome, in order to help enabling the future development of formal DfAM guidelines.

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